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<b>(54) Title:</b> PERFUME COMPOSITIONS  <b>(57) Abstract</b>  The invention concerns non-sticky, non-ethanolic fragrance formulations containing conventional odoriferous material at a level of ca. 2 %, preferably ca. 5 % to ca. 30 % which is solubilised by a combination of (a) a solubiliser and (b) a cosolubiliser in water.		

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Perfume compositions

The invention concerns non-sticky, non-ethanolic fragrance formulations containing conventional odoriferous material at a level of ca. 2%, preferably ca. 5% to ca. 30% which is solubilised by a combination of

- 5 (a) a solubiliser and  
(b) a cosolubiliser  
in water.

10 These fragrance formulations are clear, transparent or at least translucent or opaque formulations, generally solutions, or at the utmost micro-emulsions.

Suitable solubilizers (a) are in particular non-ionic surface active agents (class a<sub>1</sub>).

15 A further class (class a<sub>2</sub>) of suitable solubilizers are C<sub>1-6</sub>-alkanol propylene oxide and ethylene oxide polymers (oligomers) - and mixtures of such compounds with the non-ionic surfactants (a<sub>1</sub>).

The suitable classes are defined in detail below.

The fragrance ingredients may be of any natural and/or synthetic origin. In the foreground of the interest are (fat) soluble perfume oils.

20 (Fat) soluble perfume oils are natural or synthetic essential oils, such as orange oil, pine oil, peppermint oil, eucalyptus oil, lemon oil, clove leaf oil, cedarwood oil, bergamot oil, rosemary oil, patchouli oil, lavandin oil, spike oil, rose oil, vetiver oil, fennel oil, anise oil, thyme oil, geranium oil, lavender oil, menthol and synthetic, oil soluble  
25 perfume oils, preferably selected from the usual group consisting of alcohols, ketones, aldehydes, esters and polyenic compounds, etc. Naturally the term also encompasses any mixtures of perfume oils as defined herein. Furthermore, the term also encompasses perfume concentrates in non-ethanolic diluents as well as perfume bases in,  
30 preferably, non-ethanolic diluents.

- 2 -

The final fragrance formulations, i.e. the consumer products to which the novel formulations are diluted to with water may be in particular

5 a) water-based colognes, containing up to ca. 6 % of perfume material,

b) aqueous extrait perfumes, containing up to ca. 25%, in particular ca. 18 to 25 % perfume material,

c) aqueous Eau de Toilette containing up to ca. 18%, particularly ca. 6 to 18% perfume material.

10 It is to be noted that all figures and brackets of figures given are approximate figures.

The co-solubilizer, whose function is to promote the solubilization in a given solubilizer, is in particular a water-soluble glycol, e.g. an aliphatic glycol, such as ethylene glycol, propylene glycol, e.g. 1,2-  
15 propylene glycol, dipropylene glycol, hexylene glycol etc., or a polyethylene glycol, or a (volatile) silicone or a (volatile) silicone copolymer. The number n in the unit  $(OCH_2CH_2)_n$  of the polyethylene glycol is conveniently ca. 4 to ca. 40, e.g. 6 or 12, etc. The alkyl group of the chain can contain ether linkages.

20 At the same time, the co-solubilizer functions as an anti-sticking agent, in as far as by its use, the concentration of the solubilizer - which very frequently causes stickiness - can be lowered, e.g. to levels less than would otherwise be possible, and in particular to equal or less than 25%. Additionally the co-solubilisers (b) and in particular  
25 the silicones can by themselves improve the cosmetic touch of the formulation and reduce the stickiness.

Convenient products a) are:

30 Cremophor RH40, RH60, Tween 20, 80 (Polysorbate 20, 80), Triton X-100 (Octoxynol-9), Lamacit 877 (Nonoxynol-14) LRI (mixture of PPG 26-buteth 26 and PEG hydrogenated castor oil), Triton X 102 (Octoxynol-13) etc.

Convenient products b) are:

Dimethicone (a linear polyalkyl, i.e. a polydimethyl siloxane),

Cyclomethicone (a cyclic dimethyl polysiloxane), Phenyl Dimethicone (a polyalkylaryl siloxane), Dimethicone Copolyol (a polymer of dimethyl siloxane with polyethylene and/or polyoxypropylene side chains), Dimethiconol (a dimethyl silicone terminated with hydroxyl groups), etc.

and, in general, the entire classes to which the above commercial products belong to.

The volatile silicones such as the cyclic polysiloxanes are preferred, although also non-volatile silicones such as those polyalkyl siloxanes, polyalkylaryl siloxanes and polyether siloxane copolymers and mixtures thereof which do not exhibit an appreciable vapor pressure at ambient temperatures can be used. The preferred non-volatile silicones would be linear polyalkyl siloxanes, especially linear polydimethyl siloxanes (i.e. dimethicone).

The nature of the non-ionic surfactant (a) is not critical and practically all these surfactants may be used for the purpose of the present invention. Preferred are those surfactants with a HLB-value of 12 or greater.

As non-ionic surfactants there are, however, preferably used ethoxylated or propoxylated or propoxylated/ethoxylated derivatives of:

aliphatic alcohols, in particular C<sub>12-20</sub> aliphatic alcohols,  
alkylphenols,  
aliphatic carboxylic acid (fatty acid) esters,  
fatty acid esters of anhydrosorbitol,  
natural fats, oils and waxes.

All the various suitable types of non-ionic surfactants are well-known in the art, see e.g. the respective entries in the appropriate encyclopediae of, e.g. Ullmann, Kirk-Othmer and Römpf, as well as the appropriate cosmetic standard works, such as those from Janistyn, Nowak, Balsam/Sagarin, Shinoda, or, in particular, the CFTA International Cosmetic Ingredient Dictionary 4th ed. (The Cosmetic, Toiletry, and Fragrance Association, Washington D.C. 1991) and the various leaflets provided by the suppliers listed in the literature.

5 The term (C<sub>12-20</sub>-)aliphatic alcohols should embrace saturated and ethylenically unsaturated straight and branched, native and synthetic fatty alcohols. Examples are: cetyl, (iso) stearyl, cocoyl (i.e. fatty alcohols resulting from the hydrogenation of coconut oil), lauryl, oleyl, myristyl, capryl, tridecyl, eicosyl, alcohols.

The preferred alcohols are the C<sub>16-18</sub>-aliphatic alcohols, e.g. cetyl, stearyl and oleyl alcohol.

10 The term alkylphenol should particularly embrace mono-, di- and tri-C<sub>8-15</sub>-alkyl-phenols, whereby the alkyl radicals can be straight or branched. Examples are octyl, nonyl, dodecyl; octyl and nonyl are preferred.

15 The term aliphatic carboxylic acid (fatty acid) esters should embrace monoesters, diesters, sesquiesters of the fatty acids, e.g. of C<sub>12-18</sub>-acids, particularly C<sub>16-18</sub>-acids; examples of such acids are lauric, (iso) stearic, myristic, palmitic, stearic (preferred), linolenic linoleic, oleic, coco (i.e. fatty acids derived from coconut oil), ricinoleic, castor, etc. and hydrogenated castor oil fatty acids (preferred).

20 The term relates primarily to the respective polyoxyethylene (preferred) and polyoxypropylene/polyoxyethylene esters of the fatty acids enumerated above.

The term should, however, also embrace esters with polyhydroxy compounds, e.g. ethylene glycol, polyethylene glycol, glycerol, diglycerol, polyglycerol, propylene glycol, sorbitol, etc.

25 The term fatty acid esters of anhydrosorbitol relates to mono-, di- and triesters of sorbitan and fatty acids, e.g. to the oleates (preferred), to the laurates, to the palmitates, the stearates, etc.

The term natural fats, oils and waxes relates preferably to such compounds as castor oil (triglyceride high in ricinoleic esters), to lanolin, to beeswax, etc.

30 The average number of ethylene oxide units (degree of ethoxylation, EO) in the propoxylated and ethoxylated component (a<sub>2</sub>) is suitably between 10 and 40, preferably between 25 and 35; the

respective number of propylene oxide units (degree of propoxylation, PO) is also suitably between 10 and 40, also preferably between 25 and 35.

5 The terms "ethoxylated" and "propoxylated-ethoxylated" relate to the condensation products of ethylene oxide and/or propylene oxide with substrates having acid (movable) protons, i.e. those enumerated above for component (a<sub>1</sub>).

10 In an analogous manner, the term "propylene oxide and ethylene oxide polymer" relates to the respective condensation products of the group of compounds outlined above for a) with ethylene oxide and propylene oxide.

e.g. C<sub>1-6</sub>-alkanol +  $\begin{array}{c} \text{H}_2\text{C}-\text{CH}_2 \\ \diagdown \quad \diagup \\ \text{O} \end{array}$  condense reaction product with  
 $\begin{array}{c} \text{H}_2\text{C}-\text{CH}-\text{CH}_3 \\ \diagdown \quad \diagup \\ \text{O} \end{array}$  (in case of a<sub>2</sub>)), and

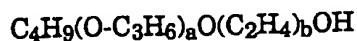
C<sub>12-20</sub>-aliphatic alcohol +  $\begin{array}{c} \text{H}_2\text{C}-\text{CH}_2 \\ \diagdown \quad \diagup \\ \text{O} \end{array}$ , or

15 e.g. C<sub>12-20</sub>-aliphatic alcohol +  $\begin{array}{c} \text{H}_2\text{C}-\text{CH}_2 \\ \diagdown \quad \diagup \\ \text{O} \end{array}$ , condense reaction product  
 with  $\begin{array}{c} \text{H}_2\text{C}-\text{CH}-\text{CH}_3 \\ \diagdown \quad \diagup \\ \text{O} \end{array}$  (in case of a<sub>1</sub>))

20 The reaction involved is generically an alkylation (i.e. an oxalkylation, a hydroxethylation), the products resulting from this reaction are thus usually termed ethoxlates or oxethylates and propoxylates or oxpropylates.

The respective figures for EO in the components a<sub>1</sub>) are suitably between 8 and 200, preferably between 30 and 50, PO is suitably 0 to 20, preferably 0 to 6, most preferably 2 to 4.

25 A preferred class of C<sub>1-6</sub>-alkanol derivatives are butanol derivatives, e.g. as represented by formula



wherein  $a = 20$  to  $40$ , preferably  $20$  to  $30$  and  $b = 20$  to  $40$ , preferably  $25$  to  $35$ .

5 If  $a = b = 26$ , the product involved is Witconol APEB (Witco Chemical, Organics Div.).

10 In all cases where a number of ethylene oxide or propylene oxide groups is given (example: butyl alcohol 26 PO 26 EO), this is an average number or molecules added. It is well-known that this number represents a statistical average, and in a given composition, the proportion of molecules present with the number shown will very often not be greater than 30 to 40% of the total number of molecules.

The preparation of the novel formulations is conveniently effected by

15 a) mixing the ethanol-free perfume concentrate and the solubilizer, preferably at room temperature until a homogeneous mixture results, and then adding the co-solubilizer, again until the mixture is homogeneous, then adding the water to such premix, slowly at first under vigorous stirring, then more quickly towards the end.

20 The premix contains conveniently from ca. 12.5% to ca. 75% fragrance material.

The solubilizer is generally more expensive than the co-solubilizer, it is thus generally indicated to work with a the minimum amount of solubilizer.

25 The novel solubilized perfume oil solutions may be used in cosmetic formulations, i.e. in aqueous solutions, in sprays, gels, etc.



- 7 -

Suitable and preferred approximate parameters are as follows:

	<u>convenient</u>	<u>preferred</u>
amount of perfume oil in fragrance solution	2% to 30%	5% to 20%
(a)	1% to 25%	5% to 18%
(b)	8% to 50%	10% to 30%
amount of a) + b)/perfume oil	1 to 10	1 to 6
amount of (a) + (b) in final fragrance solution	2% to 50%	15% to 40%
[H <sub>2</sub> O] in consumer product	20% to 95%	40% to 80%

If (b) is a (volatile) silicone, a convenient amount is ca. 0.6 - 2% and a preferred amount is ca. 0.8 to 1.5%.

5        In the context of this invention, all percentages are given on the weight basis.

Example 1

## a) Water - LRI - Propylene glycol - 15% Perfume E

	E1	E2	E3	E4	E5	E6
Perfume	15%	15%	15%	15%	15%	15%
LRI (a)	17%	17%	15.3%	15.3%	15.3%	11.9%
Propylene glycol (b)	-	25.5%	20.4%	28.9%	30.6%	34%
Water	68%	42.5%	49.3%	40.8%	39.1%	39.1%
Aspect	clear	clear	clear	clear	clear	clear
Stability	U	S	S	S	S	S
Touch	C	A	A	A	A	A

A = Cosmetically acceptable touch

5 C = Sticky

S = Stable, see also Example 2

U = Unstable

10 Sticky is defined as the skin having excessive drag when stroked with a finger after the fragrance formulation has been applied in normal conventional doses and allowed to dry.

Cosmetically acceptable is defined as the skin having an agreeable touch and is not sticky after the formulation has been applied as above.

15 Stable; the formulation is stable when it remains clear without separation into 2 or more phases when stored for a period of 1 month at temperatures in the range of ca. 5 - 45°C.

Unstable; the formulation becomes milky or separates into 2 or more phases when stored for a period of 1 month at temperatures in the range of ca. 5 - 45°C.

20 Aspect; aspect is the physical appearance at normal room temperature, either clear (transparent) or opaque (milky).

## b) Water - LRI - Propylen glycol - 20% Perfume G

	G1	G2	G3	G4	G5
Perfume	20%	20%	20%	20%	20%
LRI (a)	24%	24%	20.8%	17.6%	16%
Propylene glycol (b)	-	9.6%	14.4%	27.2%	25.6%
Water	56%	46.4%	44.8%	35.2%	38.4%
Aspect	clear	clear	clear	clear	clear
Stability	U	S	S	S	S
Touch	C	A	A	A	A

A = Cosmetically acceptable touch

5 C = Sticky

S = Stable

U = Unstable (2-phases), see Example 1

## c) Water - LRI - Dimethicone - 5% Perfum M

	M1	M3	M4
Perfume	5%	5%	5%
LRI (a)	6.65%	5.7%	5.7%
Dimethicone (b)	0.95%	0.95%	0%
Water	87.4%	88.35%	89.30%
Aspect	clear	clear	opaque
Stability	S	S	U
Touch	A	A	A

A = Acceptable, non-sticky

5 S = Stable

U = Unstable, see Example 1

## d) Water - Lamacit 877-DGP - 10% Perfume S

	S2	S4	S8	S9	S12	S11
Perfume	10%	10%	10%	10%	10%	10%
Lamacit (a)	9%	13.5%	18%	18%	9%	9%
DPG (dipropylene glycol) (b)	-	-	-	9%	23.4%	9%
Water	81%	76.5%	72%	63%	57.6%	72%
Aspect	cloudy	clear	clear	clear	clear	clear
Stability	U	U	S	S	S	U
Touch	C	C	C	A	A	A

A,C : see Example 1

5 U : unstable

\* S2, S4, S8: increase of the percentage of Lamacit : increase of the solution stability.

\* S8, S9, S12: increase of the percentage of DPG : decrease of the percentage of Lamacit.

10 \* S11, S12: increase of the percentage of Lamacit : change from a cloudy solution to a clear one.

In the above Examples Perfume E, G, M and S are conventional fragrance bases consisting of natural and synthetic ingredients, whereby these bases contain at the outmost 10% - non ethanolic -  
15 ingredients.

Example 2

The examples show that the use of (glycol) co-solubilisers allow a reduction of the amount of solubiliser required to obtain a clear solution and improve the stability of the clear solution.

5           Thus, the solubilisers (a) used in the examples 2 are :

(a) - LRI (mixture of PPG26 - buteth 26 and PEG hydrogenated castor oil).

(a) - Octoxynol 13 (eg. Triton X102.)

(a) - Polysorbate 20 (eg. Tween 20), Montanox 20

10

The co-solubilisers (b) used in the examples 2 are :

(b) - Propylene glycol,

(b) - Dipropylene glycol,

(b) - PEG 12.

- 13 -

a)

Perfume P, Solubiliser LRI, Dipropylene glycol, Water						
	P1	P2	P3	P4	P5	P6
Perfume	15	15	15	15	15	15
Solubiliser LRI (a)	18	24	28	18	18	24
Dipropylene glycol (b)	---	---	---	10	20	10
Water	67	61	57	57	47	51
Aspect	Opaque	Clear	Clear	Clear	Clear	Clear
Stability	U	U	S	U	S	S
Touch	A	C	C	A	A	A

b)

Perfume, Solubiliser LRI, Glycol co-solubiliser, Water								
	P7	P8	P10	P11	P12	P13	P14	P15
Perfume	5	5	5	5	5	5	5	5
LRI (a)	8	12	12	8	12	8	12	8
Propylene Glycol (b)	---	---	10	25	---	---	---	---
Dipropylene Glycol (b)	---	---	---	---	10	25	---	---
PEG 12 (b)	---	---	---	---	---	---	12	25
Water	87	83	73	62	73	62	71	62
Aspect	O	C	C	C	C	C	C	C
Stability	U	U	S	S	S	S	S	S
Touch	A	A	A	A	A	A	A	A

- 14 -

c)

Perfume P, Octoxynol 13, Co-solubiliser, Water					
	P16	P17	P19	P20	P21
Perfume	5	5	5	5	5
Octoxynol 13 (a)	8	10	10	10	10
Propylene glycol (b)	---	---	20	---	---
Dipropylene glycol (b)	---	---	---	30	---
PEG 12 (b)	---	---	---	---	20
Water	87	85	65	55	65
Aspect	Opaque	Clear	Clear	Clear	Clear
Stability	U	U	S	S	S
Touch	A	A	A	A	A

d)

Perfume P, Polysorbate 20, Glycol co-solubiliser, Water						
	P22	P23	P24	P25	P26	P27
Perfume	5	5	5	5	5	5
Polysorbate 20 (a)	20	25	28	20	20	20
Propylene glycol (b)	---	---	---	30	---	---
Dipropylene Glycol (b)	---	---	---	---	25	---
PEG 12 (b)	---	---	---	---	---	30
Water	75	70	67	45	50	45
Aspect	Opaque	Clear	Clear	Clear	Clear	Clear
Stability	U	U	S	S	S	S
Touch	A	A	A	A	A	A

5

The preferred Couples of (a) and (b) consist of those chemicals made use of in the Examples 1 a) to d) and 2 a) to d); hexylene glycol as a still further compound (b), must also be mentioned here.



Practical selection of solubiliser and co-solubiliser:

A suitable formulation can expediently be made as follows:

1 - A convenient solubiliser and co-solubiliser are selected, eg (a) solubiliser LRI and (b) dipropylene glycol.

5      2- The amount of solubiliser needed to approach reasonable solubilization for the required amount of fragrance oil in water is determined in the absence of co-solubiliser (b).

3 - The amount of solubiliser determined in # 2 is reduced by adding the co-solubiliser in the range as indicated.

10      4 - The levels of solubiliser (a) and co-solubiliser (b) are optimised by empirical methods so as to give a fragrance solution with the optimum aspect, stability and touch.

EP A 261 351 discloses, inter alia, stable, aqueous solutions of fat soluble perfume oils, containing

15      a) a C<sub>1</sub> to C<sub>6</sub>- alkanol propylene oxide and ethylene oxide polymer(oligomer) and

b) a non -ionic emulsifier.

20      The fragrance solutions in this reference require more solubiliser and are sticky and in general more expensive than an equivalent solution made according to the present invention, because the above mentioned component (a) does not correspond to any one of the co-solubilisers (b) of the current invention.

Claims

1. A n n-sticky, non ethanolic fragrance formulation containing conventional odoriferous material at a level of ca. 2, preferably 5% to ca. 30% which is solubilised by a combination of
- 5 (a) a solubiliser and  
(b) a cosolubiliser  
in water.
2. A fragrance formulation as claimed in Claim 1, wherein the water is present at levels between ca. 20% and ca. 95%, preferably
- 10 between ca. 40% and ca. 80%.
3. A fragrance formulation as claimed in Claim 1 or 2, in which the odoriferous material is present at levels between ca. 5% and ca. 20%.
4. A fragrance formulation as claimed in any one of Claims 1 to
- 15 3, wherein the solubiliser is present at levels between ca. 1% and ca. 25%, preferably between ca. 5% and ca. 25%, most preferably between ca. 5 and ca. 18 %.
5. A fragrance formulation as claimed in any one of Claims 1 to 4, in which the cosolubilizer is present at levels between ca. 8% and
- 20 ca. 50%, preferably between ca. 10% and ca. 40%, most preferably between ca. 10 and ca. 30 %.
6. A fragrance formulation as claimed in any one of Claims 1 to 5, in which the solubilizer is an ethoxylated derivative of a natural fat, oil or wax or an ethoxylated derivative of an phenol.
- 25 7. A fragrance formulation as claimed in any one of Claims 1 to 6, in which the cosolubilizer is a glycol or a volatile or non-volatile silicone fluid or silicone copolymer.

- 17 -

8. A premix formulation useful for preparing the fragrance formulations according to any one of Claims 1 to 7, containing conventional odoriferous material and the components (a) and (b) of Claim 1.

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Intern. Application No <b>PCT/EP 93/02555</b>		
<b>A. CLASSIFICATION OF SUBJECT MATTER</b> <b>IPC 5 A61K7/46</b>		
According to International Patent Classification (IPC) or to both national classification and IPC		
<b>B. FIELDS SEARCHED</b>		
Minimum documentation searched (classification system followed by classification symbols) <b>IPC 5 A61K</b>		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practical, search terms used)		
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US,A,4 089 814 (I. R. SCHMOLKA) 16 May 1978	1-5,7,8
Y	see the whole document ---	1-8
Y	EP,A,0 261 351 (ROURE BERTRAND DUPONT SOCIETE) 30 March 1988 cited in the application see the whole document ---	1-8
X	US,A,4 268 498 (H. GEDEON) 19 May 1981 see the whole document ---	1,3-8
X	DE,A,29 01 068 (UNILEVER N.V.) 19 July 1979 see page 21; claims 1,11,12 ---	1,2,4-8
A	DE,C,33 04 822 (B. STORP) 20 June 1984 see the whole document -----	1-8
<input type="checkbox"/> Further documents are listed in the continuation of box C. <input checked="" type="checkbox"/> Patent family members are listed in annex.		
* Special categories of cited documents :  <div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 45%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"&amp;" document member of the same patent family</p> </div> </div>		
Date of the actual completion of the international search  <div style="text-align: center; font-weight: bold;">15 February 1994</div>		Date of mailing of the international search report  <div style="text-align: center;">12.03.94</div>
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016		Authorized officer  <div style="text-align: center; font-weight: bold;">Sierra Gonzalez, M</div>

# INTERNATIONAL SEARCH REPORT

International application No.

PCT/EP 93/02555

## Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☒ Claims Nos.: 1-5, 8  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:  
The terms "solubilizer" and "cosolubilizer" are too large and may have been interpreted according to the description and examples.
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

information on patent family members

International Application No

PCT/EP 93/02555

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US-A-4089814	16-05-78	NONE	
EP-A-0261351	30-03-88	JP-A- 1052470	28-02-89
US-A-4268498	19-05-81	NONE	
DE-A-2901068	19-07-79	BE-A- 891232	16-03-82
		CA-A- 1117018	26-01-82
		CA-A- 1117424	02-02-82
		DE-A, C 2901069	19-07-79
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		JP-A- 59155254	04-09-84